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[1] Volcanic degassing produces abundant H_2O and CO_2, as well as SO_2, HCl, H_2S, S_2, H_2, HF, CO, and SiF_4. Volcanic SO_2, HCl, and H_2S have been detected from satellites in the past; the remaining species are analyzed *in situ* or using airborne instruments, with all the consequent limitations in safety and sampling, and at elevated costs. We report identification of high CO concentrations consistent with a volcanic origin (the 2010 Eyjafjallajökull and 2011 Grímsvötn eruptions in Iceland) in data from the Measurements of Pollution in the Troposphere instrument (MOPITT) onboard EOS/Terra. The high CO values coincide spatially and temporally with ash plumes emanating from the eruptive centers, with elevated SO_2 and aerosol optical thickness, as well as with high CO values in data from the Infrared Atmospheric Sounding Interferometer (IASI), onboard MetOp-A. CO has a positive indirect radiative forcing; climate models currently do not account for volcanic CO emissions. Given global volcanic CO_2 emissions between 130 and 440 Tg/year and volcanic CO:CO_2 ratios from the literature, we estimate that average global volcanic CO emissions may be on the order of ~5.5 Tg/year, equivalent to the CO emissions caused by combined fossil fuel and biofuel combustion in Australia.
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1. Introduction

[2] Volcanic gas emissions before, during, and after terrestrial eruptions commonly include H_2O, CO_2, SO_2, HCl, H_2S, S_2, H_2, HF, CO, and SiF_4 [*Symonds et al.*, 1994]; the first two constitute the largest part of volcanic emissions. The relative composition and rate of release of these volatiles contain keys to understanding the eruptive style and predicting volcanic events [*Symonds et al.*, 1994; *Thomas and Watson*, 2010]. Some volcanic gases (H_2O, CO_2, CO) have a direct or indirect positive radiative forcing and thus impact climate [*Forster et al.*, 2007].

[3] Volcanic gases were traditionally sampled *in situ* and, subsequently, in airborne campaigns. The latter are costly; both are spatially and temporally constricted as well as hazardous. Satellite detection of volcanic gases, which would be mostly free from these drawbacks, has been achieved for only a few species. Detection of volcanic water vapor and CO_2 is challenging due to high background levels of these gases in the atmosphere [*Symonds et al.*, 1994]. In contrast, due to its relative high abundance in volcanic plumes and very low background levels, volcanic SO_2 is routinely analyzed from satellite data [*Oppenheimer et al.*, 2011, and references therein]. Other volcanic species such as HCl [*Prata et al.*, 2007] and H_2S [*Clarisse et al.*, 2011] have also been successfully identified from satellites.

[4] To test the feasibility of volcanic CO detection from satellite we have focused our efforts on the analysis of several datasets acquired over the Iceland region (58°N, −28°E to 68°N, −10°E) during the Eyjafjallajökull 2010 and Grímsvötn 2011 eruptions.

2. Observations

[5] In this study we use satellite observations acquired by the MOPITT, MODIS, IASI, and OMI instruments. MOPITT is a nadir-looking, cross-track scanning infrared radiometer onboard EOS/Terra that uses gas correlation spectroscopy to detect CO in the troposphere. We used version 4, level 2 MOPITT CO retrievals obtained from its thermal infrared (TIR) channels (7D, 5D, and 5A; D and A channels are sensitive to target-gas and background information, respectively). These channels sense incoming radiation in the 4.56 to 4.67 μm spectral range, coinciding with the R-branch of the CO rotational-vibrational fundamental mode. MOPITT provides total CO column values as well as some information on CO vertical distribution. Global MOPITT coverage is achieved in approximately 3 days; its ground instantaneous field of view (GIFOV) is 22 by 22 km^2 at nadir. We used day-only, cloud-free, otherwise unfiltered MOPITT data. To maximize the number of MOPITT retrievals, cloud detection was based on MOPITT TIR radiances only; this did not distort the extent of the plumes or their retrieved CO values. MOPITT data were corrected for a systematic geolocation error by shifting the reported longitude 0.35° to the east (web3.acd.ucar.edu/mopitt/GeolocationBiasReport.pdf).

[6] To locate volcanic plumes and contrast them with MOPITT CO values we inspected simultaneously acquired EOS/Terra MODIS (Moderate Resolution Imaging Spectroradiometer) true color images with an effective GIFOV of 0.5 by 0.5 km^2 at nadir. EOS/Terra MODIS aerosol optical thickness (AOT) at 0.55 μm was also analyzed; level 2 products with a 10 by 10 km^2 GIFOV from version 051 processed after 28 September 2010 (thus not affected by an error involving incorrect clear sky radiances) were utilized.

[7] IASI total CO column data (level 2, version v20100815) for dates with anomalous MOPITT CO values and visible

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volcanic plumes were also analyzed. MOPITT and IASI apply different measurement techniques and retrieval algorithms to derive CO abundances, thereby offering two independent sets of evidence to investigate the detectability of volcanic CO from satellite. IASI is a nadir looking Fourier transform infrared spectrometer onboard MetOp-A. It covers the 3.6–15.5 μm spectral range sampling every 0.25 cm^{-1}, and thus can resolve individual absorption lines of CO and other atmospheric infrared active species. Global coverage is achieved twice daily with a 12 km diameter GIFOV at nadir. IASI retrievals are commonly filtered for clouds based on Eumetsat cloud information; additionally, we used coeval MetOp-A AVHRR/3 data to ensure that only cloud-free IASI pixels were utilized in this analysis. We also analyzed IASI brightness temperature (BT) spectra in the MOPITT TIR channel region to derive the depth of diagnostic CO lines, which is related to CO abundance, and to investigate the radiative effects of volcanic aerosols.

[8] SO_2 is a well established atmospheric volcanic marker. We analyzed total SO_2 column values (data set version 003, level 2) obtained from measurements by the Ozone Monitoring Instrument (OMI) onboard EOS/Aura. OMI provides daily global coverage with an GIFOV of 24 by 13 km^2 at nadir. For each OMI scene lower troposphere (TRL) and mid troposphere (TRM) operational SO_2 column density values are provided; these values are obtained assuming a center of mass altitude (CMA) for the volcanic plume at 2.5 and 7.5 km, respectively. Column density values for CMA between these can be obtained by linear interpolation (http://so2.gsfc.nasa.gov/Documentation/OMSO2Readme_V111_0818.htm). We discarded pixels with radiative cloud fraction ≥0.2.

[9] The altitude of the volcanic cloud during the 2010 eruption was constrained using measurements from the C-band weather radar located in Keflavík International Airport [*Arason et al.*, 2011]. A report describing the 22 May 2011 eruption contains plume height estimates based on similar measurements [*Jakobsdóttir et al.*, 2012].

3. Results

[10] We have identified clusters of anomalously high MOPITT CO values on 19 April 2010 and 7 May 2010 (Eyjafjallajökull eruption), as well as on 22 May 2011 (Grímsvötn eruption). We consider anomalous values the highest retrievals (on a specific date, in the study area) which coincide spatially with a volcanic plume in simultaneously acquired MODIS visible data. Similarly, we consider background values the retrievals outside that plume. Anomalous and background values represent two separate populations, according to their value and spatial distribution.

[11] The anomalous MOPITT CO values on the three dates are spatially continuous and have a plume-like distribution (Figure 1, left). The reported average height for the top of the volcanic plume during satellite overpass was 2.5 km (19 April) and 5.3 km (7 May) [*Arason et al.*, 2011]. On 22 May the lower boundary of the volcanic plume to the south of the vent was at about 5 km [*Jakobsdóttir et al.*, 2012]. Compelling evidence of further anomalous values during the remaining dates of volcanic activity in 2010 and 2011 was not found due to either meteorological clouds precluding the CO retrievals or to lack of sufficient MOPITT coverage. We calculate the difference between MOPITT total CO column in the plume versus that in the background to be between 4.48 and 4.05e + 017 mol/cm^2 in the Eyjafjallajökull eruption and 1.89e + 017 mol/cm^2 in the Grímsvötn eruption (Table S1 in the auxiliary material).[1]

[12] IASI CO total column data on 7 May 2010 show a plume-like set of anomalously high values which coincide spatially with these shown by MOPITT. Their mean exceeds the average CO background value by 6.38e + 017 mol/cm^2 (Figure 2 and Table S1). IASI data on 19 April 2010 and 22 May 2011 over the plumes were deemed cloudy according to the Eumetsat cloud information (i.e., cloud coverage in the pixel ≥12%), and thus sufficient spatial coverage for these two dates is not available.

[13] On the three dates analyzed the anomalous MOPITT CO values show good overlap with volcanic plumes apparent in the MODIS visible images and with high AOT values (Figure 1, middle). MODIS measured a wide range of AOT values over the MOPITT CO plumes: 0.2 to 2.96 (19 April 2010), 0.09 to 1.54 (7 May 2010), and 0.001 to 2.7 (22 May 2011).

[14] OMI shows SO_2 plumes emanating from the volcanic centers on all three dates (Figure 1, right). We have derived SO_2 column values by linear interpolation between operational OMI TRL and TRM retrievals, for a CMA equal to the reported height of the top (or bottom, depending on availability) of the volcanic cloud during satellite overpass. By using the height of the cloud's top rather than the center of mass altitude the SO_2 column values are slightly underestimated (~8.5%, for a 2 km thick cloud). Using the clouds' bottom results in a similar overestimation. We calculate an average SO_2 total column value of 5.76e + 017 (plume directly south of the vent on 19 April 2010), 1.12e + 018 (7 May 2010), and 3.75e + 018 mol/cm^2 (22 May 2011), or 2.14, 4.17, and 13.96 DU (Dobson Units), respectively. These measurements are well above OMI's detection limit [*OMI Team*, 2012].

4. Discussion

4.1. Artifact or Volcanic CO?

[15] In order to determine if the anomalous MOPITT CO values in the volcanic plumes is an artifact produced by other volcanic gases and/or volcanic aerosols we have investigated other sources of absorption in the MOPITT TIR bandpass. Due to the nature of gas correlation radiometry, non-target gases have very little effect on the measured radiances. Additionally, most relevant volcanic gases have no strong absorption lines in the MOPITT TIR bandpass (e.g., CO_2, SO_2, HCl, H_2S, S_2, H_2, HF, SiF_4, SO, N_2, CH_4, Ar, O_2, and NH_3). The opposite is true for water vapor and carbonyl sulfide (a minor volcanic gas); however, the latter is nearly two orders of magnitude less abundant than CO in average (from data in *Symonds et al.* [1994]) and thus would produce a much weaker signal.

[16] To model the sensitivity of MOPITT CO retrievals to elevated water vapor we used MOPABS (the MOPITT absorption model [*Edwards et al.*, 1999]). We ran simulations with a standard atmosphere input and then modified that input to include: 1) a CO profile increased by 10% at all pressure levels, 2) the average background NCEP/GDAS

[1] Auxiliary materials are available in the HTML. doi:10.1029/2012GL053275.

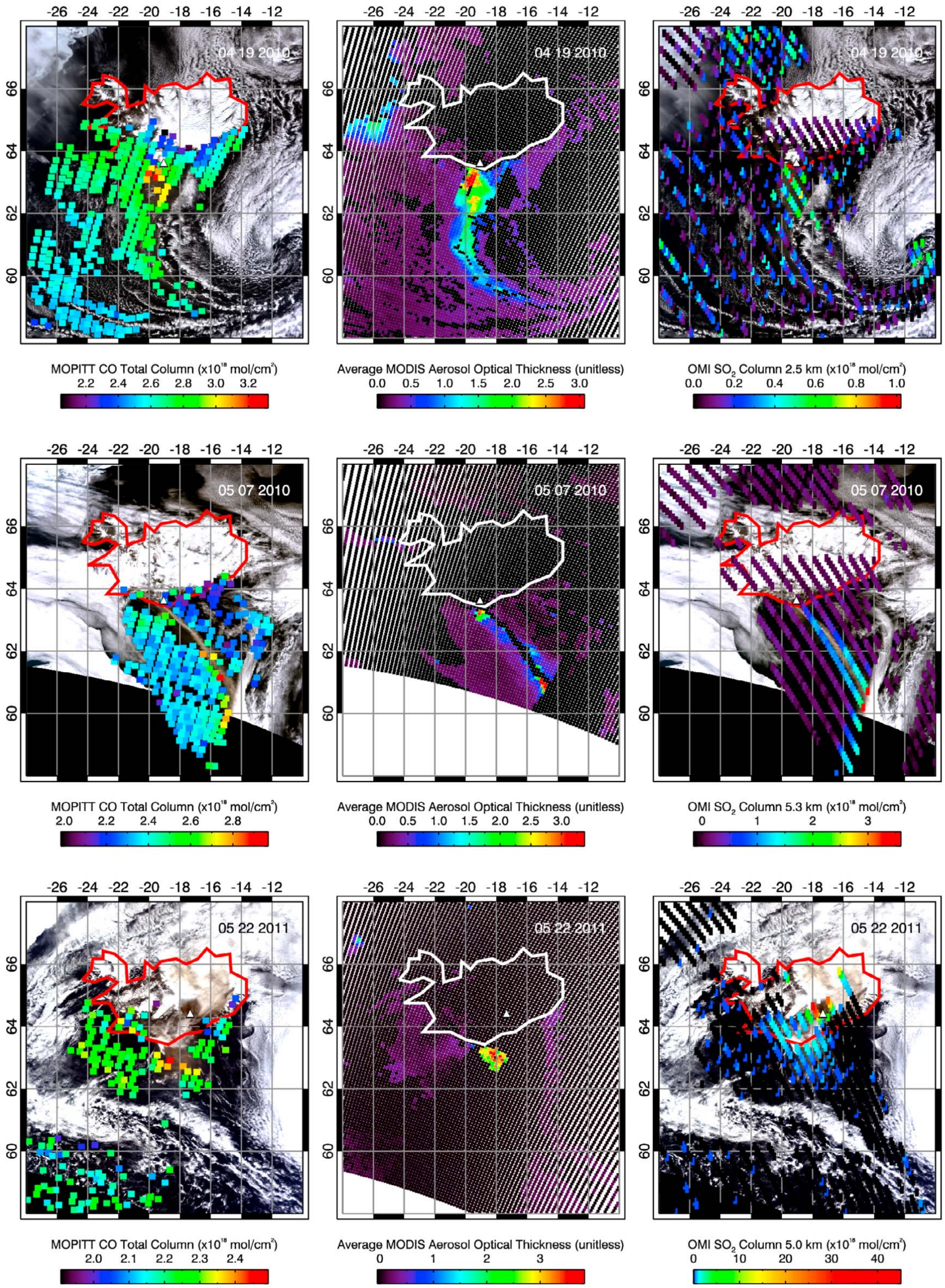

Figure 1. Volcanic markers mapped for the three dates analyzed. Iceland's contour and the location of the active volcanic vent (white triangle) are shown for clarity. The size of the colored pixels is that of the instrument's GIFOV, unless noted otherwise. (left) MOPITT total CO column values over coeval MODIS true color image. Anomalously high CO values over the volcanic plume appear in yellow and red. (middle) MODIS AOT. Plume values appear in cyan, green, yellow, and red. (right) OMI total SO_2 column over coeval MODIS true color image. Anomalously high SO_2 values appear in cyan, green, yellow, and red. OMI pixels shown enlarged (×2) for clarity.

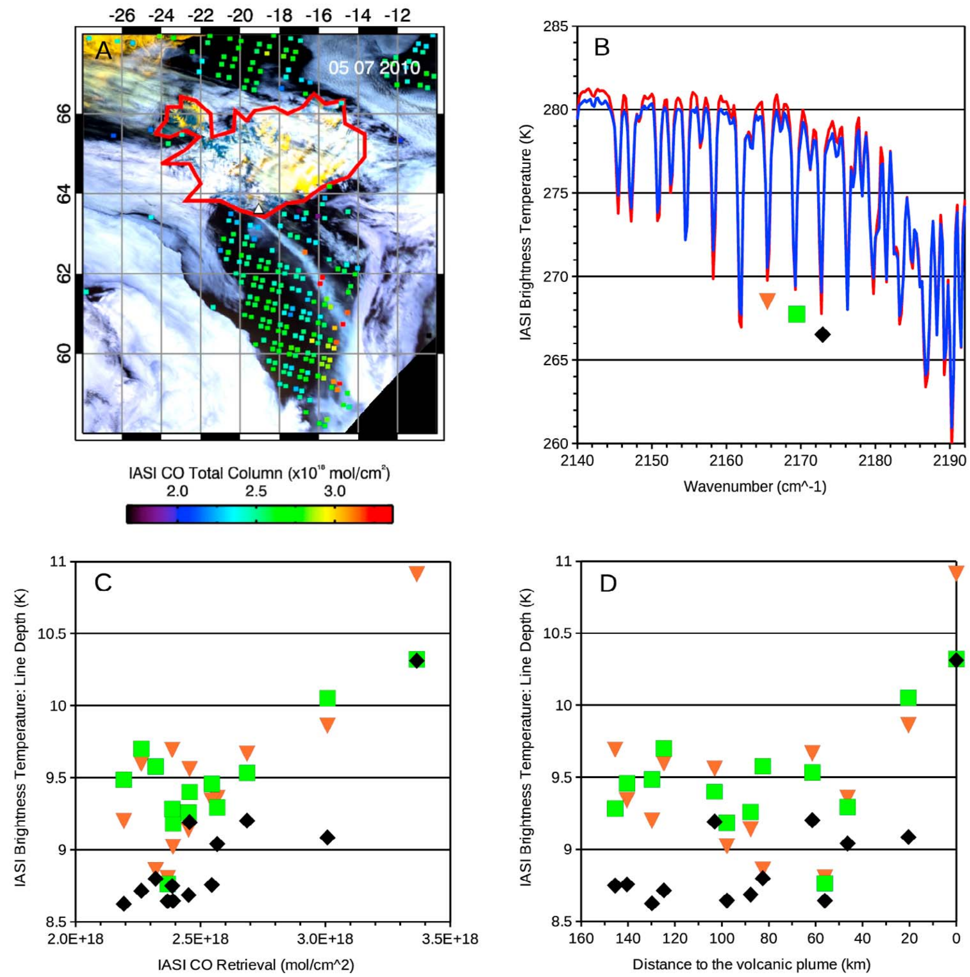

Figure 2. (a) IASI total CO column values on 7 May 2010 over coeval AVHRR/3 image. Anomalously high CO values in the volcanic plume appear in orange and red. IASI scanlines run west-northwest to east-southeast. (b) IASI BT spectra acquired in one scanline inside (red) and away from (blue) the volcanic plume. The spectral range shown is that of the MOPITT TIR channels; selected CO absorption lines (at 2165.5, 2169.2, and 2172.7 cm^{-1}) are indicated with colored symbols. Slight BT differences between spectra continuum (less than 1 K in the example shown) are not systematic and could be due to meteorological effects, among others. (c) Depth of CO lines in IASI BT spectra acquired in one scanline versus IASI CO retrievals (symbols as in Figure 2b). (b) Depth of CO lines in IASI BT spectra acquired in one scanline versus distance to the volcanic plume (symbols as in Figures 2b).

(National Centers for Environmental Prediction/Global Data Assimilation System) water vapor profile for 7 May 2010, and 3) the average NCEP water vapor profile for the plume locus on that same date. (NCEP/GDAS water vapor profiles, which are utilized in operational MOPITT CO retrievals, are derived from multi-sensor measurements; climatological data are utilized to complement lacking or insufficient measurements. We have corroborated that water vapor profiles in the volcanic plume and its surroundings do not include climatological data.) Modeling results (Table 1) show that the change in MOPITT TIR D signal due to increased water vapor in the plume is 1 to 2 orders of

Table 1. MOPABS Sensitivity Test Results: Effect on MOPITT TIR Radiances of a 10% CO Increase (the MOPITT Detection Limit) Versus That Produced by Differences in Water Vapor Between the Volcanic Plume and Its Surroundings[a]

	MOPABS Radiance (W m^{-2} sr^{-1})		
	MOPITT TIR Channel 7D	MOPITT TIR Channel 5D	MOPITT TIR Channel 5A
Standard atmosphere	2.11064E-4	1.31012E-2	4.55677E-2
10% CO enriched atmosphere	2.02425E-4	1.30718E-2	4.55336E-2
Water vapor profile: background	2.11615E-4	1.31502E-2	4.57532E-2
Water vapor profile: plume	2.11579E-4	1.31450E-2	4.57294E-2
D/A change: standard vs CO enriched	4.09%	0.22%	-
D/A change: background vs plume	0.02%	0.04%	-

[a]Water vapor profiles are NCEP averages for 7 May 2010.

magnitude below that due to a 10% change in CO, which is the MOPITT detection limit [*Pan et al.*, 1995].

[17] Volcanic aerosols are strong absorbers at longer wavelengths (i.e., near 10 μm) [*Clarisse et al.*, 2010]; their behavior in the MOPITT TIR bandpass is less well understood. To investigate this point we analyzed IASI BT spectra on 7 May 2010, focusing on sets of cloud-free, adjacent spectra acquired in the same scanline inside of and away from the volcanic plume. Our analysis shows that in the spectral region of the MOPITT TIR bandpass, aerosol-free and aerosol-rich spectra (from background and plume, respectively) are indistinguishable in terms of the shape of their continuum and individual absorption lines (Figure 2b). Three of the diagnostic CO absorption lines in this spectral range (at 2165.5, 2169.2, and 2172.7 cm-^{1}) were selected for further analysis, based on their intensity and lack of overlap with H_2O lines. Their depth was quantified and contrasted to IASI CO values and distance to the volcanic plume. As expected, line depth increases with CO abundance (Figure 2c). For distances less than 40 km, line depth and thus CO abundance are inversely proportional to distance between the spectrum locus and the plume (Figure 2d). For distances greater than this threshold line depth and distance show no correlation. This behaviour of the CO spectral features with respect to proximity of the plume is consistent with a volcanic origin for the anomalously high CO values in the plume.

4.2. Satellite Detection of Volcanic CO Versus Aircraft Observations and Other Volcanic Markers

[18] The maximum MOPITT and IASI CO values in the plume differ in magnitude by less than 14% (2.97e + 018 and 3.37e + 018 mol/cm^2, respectively, on 7 May 2010). MOPITT mixing ratio profile values are also close in magnitude to airborne CO measurements during the 2010 eruption (CARIBIC project [*Brenninkmeijer et al.*, 2007] and DLR-Falcon campaign [*Schumann et al.*, 2011]). CARIBIC flights on 20 April, 16 May, and 19 May 2010 intersected the volcanic plume at an approximate altitude of 4 km (600 hPa) over northern Germany, Northern Ireland and the Norwegian Sea, respectively. The average CO background values were near 120 ppbv, while values in the plume for each of the three dates reached 200, 205, and 179 ppbv respectively [*Rauthe-Schöch et al.*, 2012]. The flights in the DLR-Falcon campaign sampled a fresh plume between Iceland and northern Scotland on 2 May at 3.5 km altitude (661 hPa), with background and plume CO values of 128.5 and 200.9 ppbv, respectively. Despite the spatial and temporal differences, we find that in most cases airborne CO measurements and MOPITT mixing ratio values derived for similar pressure levels (Table S1) agree within 10%.

[19] As shown by OMI measurements, volcanic SO_2 plumes are present on all three dates analyzed and coincide spatially with the MOPITT and IASI CO plumes, as well as with high MODIS AOT values. On 19 April 2010 a second cluster of anomalously high SO_2 values was detected away from the vent, near 59°N, -11°E; this would be consistent with an older volcanic cloud transported SE by the prevalent winds. We find that average SO_2 total column in the plume is roughly of the same magnitude (19 April 2010) or one order of magnitude above (7 May 2010 and 22 May 2011) the average MOPITT CO total column values. This is consistent with CO:SO_2 ratios obtained from field and airborne measurements in volcanic environments where tectonic plates diverge, such as Iceland [*Symonds et al.*, 1994; *Oppenheimer et al.*, 2011].

5. Summary and Conclusions

[20] We report satellite identification of volcanic CO for the first time, achieved using data from two independent instruments: MOPITT and IASI. Other volcanic markers (ash plume in MODIS visible images as well as elevated MODIS AOT and OMI SO_2) are spatially and temporally co-located with the anomalous CO values and thus confirm their volcanic origin.

[21] We have ruled out a spurious origin for the anomalous CO values due to water vapor or aerosols in the volcanic plume. By modeling the effect of water vapor in MOPITT radiances we have shown that differences between water vapor observed in the plume versus the background could not account for the change in measured MOPITT radiance. Our analysis of IASI BT spectra indicates that the MOPITT TIR bandpass is not affected by aerosols in the volcanic plume. We have also shown that the depth of diagnostic CO lines increases as the distance to the volcanic plume decreases. Since the depth of these CO absorption lines should increase with increasing CO, we interpret this as definitive evidence that the high CO values retrieved in the plume are volcanic in origin.

[22] CO emissions result in an increase of CO_2, CH_4, and tropospheric O_3, and thus have a positive indirect radiative forcing of approximately 0.2 W/m^2 (i.e., 12.5% of the total net anthropogenic forcing) [*Forster et al.*, 2007]. Global CO emissions measured in the 2002–2009 period range widely, between 1318 and 1504 Tg/year [*Fortems-Cheiney et al.*, 2011]; it is assumed that they are dominated by incomplete fossil fuel combustion and biomass burning. Fluctuations in gas emissions from natural sources (e.g., volcanic degassing) need to be quantified so as to separate them from those of anthropogenic origin.

[23] The magnitude of global volcanic CO emissions is unknown. Next, we estimate their approximate value to put them into perspective respect to other CO sources. Global volcanic CO_2 emissions have been estimated to be between 130 and 440 Tg/year (from data in *Gerlach* [2011]). Volcanic CO and CO_2 emissions depend on tectonic environment and may vary from eruption to eruption. However, and unlike CO and SO_2, we find that CO and CO_2 are correlated to some degree, regardless of tectonic setting. Based on mean global volcanic CO_2 and linear regression parameters derived from volcanic CO and CO_2 measurements [*Halmer et al.*, 2002; *Wardell et al.*, 2004; *Oppenheimer et al.*, 2011] we estimate that global average volcanic CO emissions may be on the order of $\sim$5.5 Tg/year; errors from the linear regression are negligible compared to the CO_2 estimates. This is equivalent to the annual CO emissions produced by fuels (fossil and biofuel) in Australia [*Kopacz et al.*, 2010]. Though modest compared to anthropogenic sources, volcanic CO emissions are non-negligible.

[24] Satellite measurements of volcanic CO may lead to a better understanding of variations in the global CO budget, and thus improve climate models. They will also refine our understanding of volcanic processes by answering basic questions such as how much CO is emitted in an eruption

and what is the relative gas composition in different volcanic events.

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